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AFOSR Final Scientific Report

Mechanisms of Initiation of Detonation in Explosive Vapor Clouds

by

R. Knystautas and J.H. Lee

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ABSTRACT

During this period emphasis has been placed mostly on problems pertaining to feasibility of FAE III weapons. In particular major efforts have been devoted to the study of the mechanisms of initiation using various techniques other than the blast initiation method via concentrated explosive charges of conventional FAE weapons. Perhaps the most significant progress is the discovery of the universal initiation mechanism of Shock Wave Amplication by Coherent Energy Release (SWACER). When conditions for the SWACER mechanism to work have been generated locally in the explosive gas, then initially weak shocks can amplify extremely rapidly to form detonations in a time scale of the order of microseconds and corresponding length scales of the order of centimeters for the sub-atmospheric fuel-oxygen mixtures studied. It has been demonstrated experimentally that flash photolysis, intense turbulent mixing of either a pyrophoric compound (DMZ) or hot combustion products with the explosive gas can lead to direct initiation via the SWACER mechanism. Theoretical modelling of the SWACER mechanism is being performed to achieve the necessary criteria for scaling up the present laboratory scale experiments to field tests and to the use of fuel-air mixtures rather than the more detonable fuel-oxygen mixtures of the present work.

Introduction

The present report gives a comprehensive summary of the
results of a number of studies of the non-blast mode of detonation
initiation in explosive gas media. In this mode of initiation one
does not rely on the blast wave from a powerful source to initiate
the detonation. Three separate studies were carried out in which
direct initiation of detonation in an explosive gas was achieved
photochemically by the UV production of free radicals, catalytically
by the dispersion of a pyrophoric material and finally via a turbu-
lent hot gas jet. The present work is deemed of great potential
relevance to the development of single event third generation FAE
weapons.

The most significant result was the discovery of a very
rapid shock amplification mechanism which is active when the right
conditions are created in the explosive gas medium. This mechanism
has since been called the SWACER mechanism (for $\underline{S}$hock $\underline{W}$ave $\underline{A}$mplifi-
cation by $\underline{C}$oherent $\underline{E}$nergy $\underline{R}$elease) and is now believed to be univer-
sal whenever detonation is initiated without a source-created blast
wave. Unlike the blast mode of detonation initiation via a power-
ful source, such as that used in second generation FAE devices, the
SWACER mechanism relies on the exothermic energy release of the ex-
plosive medium itself to produce detonation very quickly after the
triggering of chemical reactions. The essential requirement for
the SWACER mechanism to be operative is that a suitably tailored
gradient of induction time be produced in the explosive gas medium.
When the explosive gas is so conditioned then the pressure waves
produced when chemical reactions are triggered are amplified very
rapidly from acoustic to detonation levels. The appropriate gradient

in induction time of the explosive mixture can be produced in a
number of ways, for example: non-thermally via a free radical
concentration gradient produced photochemically or by the dispersal
of a catalytic compound or thermally via a temperature gradient
in the mixing layer between an injected hot turbulent jet and the
quiescent explosive gas mixture. In a typical sub-atmospheric
hydrocarbon fuel-oxygen explosive gas medium photochemically seeded
with free radicals, times for the amplification of pressure waves
to detonation levels of the order of tens of microseconds have been
achieved. Similar amplification rates have been produced by using
pyrophoric materials or a hot turbulent jet as the initiating agents.

Since some of this work has already been presented at
scientific conferences and is or soon will be in various stages of
publication, each initiation technique is discussed in this report
as a self-contained Appendix.

Appendix I

Photochemical Initiation of Gaseous Detonations*

Photochemical Initiaton of Gaseous Detonations

By

John H. Lee, R. Knystautas and N. Yoshikawa
Department of Mechanical Engineering
McGill University, Montreal, Canada

ABSTRACT

Direct initiation of detonations in gaseous mixtures of C_2H_2-O_2, H_2-O_2 and H_2-Cl_2 in the pressure range of 10 to 150 torr using flash photolysis was studied. Similar to blast initiation using a concentrated powerful energy source, it was found that for photochemical initiation, there exists a certain threshold of flash intensity and energy for each mixture at any given initial pressure and composition below which a deflagration is formed. At the critical threshold, however, a fully developed detonation is rapidly formed in the immediate vicinity of the window of incident UV radiation. However, at super critical flash energies, the amplitude of the detonation formed decreases and combustion of the entire irradiated volume approaches a constant volume explosion. It was found that photo-chemical initiation requires both a certain minimum peak value of the free radical concentration generated by the photo-dissociation as well as an appropriate gradient of this free radical distribution. The minimum peak radical concentration permits rapid reaction rates for the generation of strong pressure waves, while the gradient is necessary for the amplification of the shock waves to a detonation. If the gradient is absent and the free radicals are uniformly distributed in the mixture, then the entire volume simply explodes as in a constant volume process. The present study reveals that the mechanism of photochemical initiation is one of proper temporal synchronization of the chemical energy release to the shock wave as it propagates through

the mixture. In analogy to the LASER, the term SWACER is introduced to represent this mechanism of Shock Wave Amplification by Coherent Energy Release. There are strong indications that this SWACER mechanism is universal and plays the main role in the formation of detonations whenever a powerful concentrated external source is not used to generate a strong shock wave in the explosive.

1. Introduction

To initiate a detonation wave in an explosive mixture, a powerful concentrated energy source (eg., a solid explosive charge, electric sparks, exploding wire, etc.) is usually employed. A very strong blast wave is generated by the source which initiates chemical reactions in its wake. If sufficient energy is used to give the blast a long enough duration as well, then direct initiation is achieved. Thus in blast wave initiation, a strong shock is always generated by the external energy source. Successful initiation depends on whether or not sufficient source energy is used to give the blast wave a long enough duration before it decays to a strength below the auto-ignition li... The mechanisms of blast initiation are now fairly well understood and theorie [1,2] are available to give quantitative predictions of the blast energy required for direct initiation from the properties of the explosive mixture (eg. induction times, equilibrium Chapman-Jouguet properties, etc.).

In considering the important question of the possibility of transition from deflagration to detonation of an unconfined vapor cloud, one is faced with the problem of discovering what mechanisms are available for generating strong shocks of sufficiently long duration. Without an external concentrated energy source, it is not at all obvious how solely on the basis of the energy release from the combustion of a mixture in a turbulent

flame that shock waves of the order of $M_s \approx 4$ or 5 and duration of the order of few tens to few hundreds of microseconds (corresponding to common hydrocarbon-oxygen and air mixtures, respectively) can be obtained even under confined conditions. In an effort to elucidate these mechanisms of "self-initiation" (a term proposed for initiation·without the use of powerful high energy external sources[3]) an investigation was carried out on photo-chemical initiation of detonation. The present paper reports the results of this study.

In flash photolysis, chemical reactions are initiated by irradiating an explosive gas mixture by a strong flash. Photo-dissociation produces a distribution of free radicals in the mixture which then initiates the chain and branching reactions leading to the explosion of the mixture itself. Although a large quantity of energy is used to produce the flash, the actual amount of light energy absorbed by the mixture is relatively small and the initiation of chemical reactions is of a non-thermal nature with the free radicals responsible for starting the reactions. Flash photolysis is a well-established means of studying homogeneous gas phase reactions[4] and photo-chemical ignition and combustion enhancement of fuel-oxidizer mixtures were investigated by Cerkanowicz[5]. It appears that if a large concentration of free radicals is generated by the flash, the subsequent reactions can be sufficiently rapid to cause detonation waves to form in the irradiated gas mixture. Detonation-like phenomena in a reaction tube have been reported by Thrush[6] and Wadsworth[7] who appear to be the first to use flash photolysis for the direct initiation of NO_2 sensitized C_2H_2-O_2 cylindrical imploding detonations. The detailed mechanisms of direct initiation by the free radicals generated via flash photolysis have not been analyzed by Thrush and Wadsworth. The present study attempts to reveal these mechanisms more explicitly via photographic observations.

2. Experimental Details

Three apparatii have been employed in the present study. The first apparatus (Fig. 1) is in essence a double elliptical laser cavity "pumped" by a pair of xenon flash tubes (EG&G FX-47C). In place of the neodymium rod in the laser cavity, a quartz tube of 42.5 cm long, 14 mm I.D. and 1 mm in wall thickness was placed in the center of the cavity between the two xenon flash tubes. The cavity is 16 cm long and the quartz tube extends 120 cm outside the laser caivty to permit observation of the combustion phenomena outside the 16 cm irradiated portion of the quartz tube inside the cavity. At the cavity end of the quartz tube, a piezoelectric transducer (PCB 113A24) was placed to record the pressure. The flash tubes and the quartz explosion tube were water-cooled and the cavity was flushed with dry nitrogen. The combustion wave outside the cavity was recorded by a high speed streak camera. A photo-tube (RCA 929) was used to monitor the light output of the flash tubes so that various events could be referenced to the start of the light emission inside the cavity.

To permit schlieren photography of the initiation processes, a second apparatus (Fig. 2) was used. This consisted of a rectangular box 75 mm x 75 mm of square cross sections and 150 mm long. The four sides of the box were constructed using UV transmitting quartz plates 75 mm x 150 mm and 6 mm thick, while the two ends were closed by stainless steel plates with a centrally mounted pressure transducer to record the explosion pressure. Two xenon flash tubes (Xenon Corp. FPA-8-100C) were placed adjacent to two opposite quartz plates to irradiate the explosion chamber. Two parabolic reflectors were placed between the quartz tubes to enhance the irradiation. The schlieren photography was taken through the other pair of quartz windows orthogonal to the direction of the UV irradiation. Due to the presence of the frames in which the quartz windows were mounted, the total unobstructed schlieren

field was only 50 mm x 12 cm and about 10 mm of the gas from the quartz plates were not visible for schlieren observations.

In subsequent experiments, it was found that detonation formation occurs within the first 10 mm in the immediate vicinity of the quartz windows of the incident UV radiation. This necessitated a third apparatus to be constructed if the region immediately adjacent to the window of incident UV radiation were to be observed. The third apparatus (Fig. 3) consisted of a cylindrical chamber 100 mm I.D. and 100 mm long, and the ends were closed by two 100 mm diameter quartz windows to permit schlieren observation of the entire explosion chamber. A 35 mm diameter quartz window was installed on the side at right angles to the axis of the cylindrical chamber and the UV radiated through this window was from a xenon flash tube placed 30 mm behind this window. In this apparatus an edge-on view of the window of incident UV radiation could be obtained and the entire phenomena occuring at the window surface could be observed photographically.

Premixed mixtures of $C_2H_2-O_2$, H_2-O_2 and H_2-Cl_2 were prepared in steel storage tanks and allowed to mix by diffusion for 24 hours prior to the experiments. Nitrogen dioxide was used in certain experiments to enhance the absorption of $C_2H_2-O_2$ and H_2-O_2 mixtures. Due to the high UV absorption of chlorine, initiation could be achieved in H_2-Cl_2 mixtures with relatively low flash intensities. Thus, within the scope of the present study of establishing the initiation mechanisms, mostly H_2-Cl_2 mixtures were used throughout the experiments to avoid "hard pumping" and this prolonged the life of the flash tubes.

3. Experimental Results and Discussions

Using Apparatus 1, direct initiation of detonation in all the mixtures

$(C_2H_2-O_2, H_2-O_2, H_2-Cl_2)$ tested in the pressure range of 10 to 150 torr could be achieved. Using the start of the flash as the reference time $t = 0$, a sudden pressure rise was recorded by the pressure transducer at the ignition end of the quartz tube after a delay of the order of a few hundred microseconds. The pressure rise is sharp and its magnitude corresponds to about the reflected C-J pressure of the mixture used. This indicates that a fully developed detonation is already formed within the 16 cm of the irradiated length of the quartz tube inside the flash cavity. A typical pressure trace superimposed on the photo tube output recording the light of the flash tubes is shown in Fig. 4. The half width of the flash is about 1 millisecond and in the particular experiment shown, the detonation is formed in the $C_2H_2 + O_2$ mixture sensitized by 5% addition of NO_2 with a delay of 550 microseconds. Simultaneous recording by the streak camera also shows that a fully established detonation emerges from the flash cavity after the delay time as registered by the pressure transducer. No pre-detonation flame is observed indicating that the detonation must have been formed inside the cavity itself. The induction time (as measured from the beginning of the flash to the appearance of the explosion) is found to depend on the particular mixture used, its initial pressure and composition, as well as the flash energy and NO_2 concentration (for the cases where NO_2 is used for enhancing photo-absorption). The results are shown in Fig. 5 and indicate the expected general behavior that the induction delay decreases with i) increasing initial pressures, ii) increasing NO_2 concentration and iii) increasing flash energy. The H_2-Cl_2 system has the minimum induction delay, while the H_2-O_2 system has the maximum. With NO_2 sensitization, direct initiation in C_2H_2-air mixture can also be achieved with sufficiently high flash energy; direct initiation in $C_2H_2-O_2$ and H_2-O_2 systems can be achieved without the use of NO_2 as sentizer. The induction times for all the experiments performed are all of

the order of a few hundred microseconds to a millisecond.

The kinetics of photo-initiation of explosion in gaseous mixtures of C_2H_2-O_2, H_2-O_2 and H_2-Cl_2 are well known and need not be discussed here again. With a sufficiently high concentration of free radicals generated by photo-dissociation, it is expected that the subsequent reactions are sufficiently rapid to result in detonation formation. Since both the flash tubes and the quartz explosion tube inside the flash cavity are water cooled, thermal effects are of negligible importance and the initiation of the chemical reactions is of a non-thermal nature via the free radicals. With the quartz explosion tube of only 7 mm radius and the photon mean free path of order of about 5 mm for sensitized C_2H_2-O_2 and 25 mm for H_2-Cl_2 under the conditions of the present experiments, the excitation of the irradiated volume can be considered homogeneous. It is not clear however, how a homogeneous volume explosion can initiate a detonation. Assuming a constant volume explosion of the 16 cm length of the excited volume inside the cavity, the pressure rise is of the order of ten times the initial pressure. From one-dimensional shock tube theory, the strength of the shock wave produced in the section outside the cavity is only about $M_2 \simeq 2$ which is not even sufficient to cause auto-ignition of the mixture. Thus it appears that the homogeneous constant volume explosion shock tube mechanism cannot explain the present results. Detonation initiation must occur locally inside the excited volume itself.

To elucidate the initiation mechanisms, a series of schlieren photographic observations were made in Apparatus 2. Typical schlieren records with the corresponding pressure records from the two pressure transducers at the ends of the rectangular box are shown in Fig. 6. To prove that shock waves result only from the combustion of the mixture,

a pure Cl_2 mixture was irradiated (Fig. 6a). Neither the schlieren records nor the pressure transducers reveal the generation of pressure waves. With an explosive mixture (H_2-Cl_2 at p_0 = 100 torr) it was found that when the flash intensity reaches a certain threshold value, direct initiation is achieved and two fully established detonation waves emerge from the top and bottom irradiated walls of the box into the schlieren field (Fig. 6b). Initiation is expected to occur at the walls first because the free radical concentration is highest there where the UV radiation enters. The corresponding pressure records indicate that the detonation pressures are about the C-J value of the mixture. The first signal of the oscilloscope trace registers the flash light output, the break in the trace denotes the time when the schlieren is taken and the pressure signal appears last. With increasing flash intensity, the induction time decreases, the amplitude of the detonation pressure drops slightly but little change in the detonation velocity can be observed (Fig. 6c). However, for very high flash intensity, the wave amplitude becomes much weaker and the explosion pressure approaches about half the C-J detonation pressure corresponding to a constant volume explosion inside the box (Fig. 6d). For even higher flash intensities, no distinct pressure waves can be recorded in the schlieren photographs and the pressure gauges record the constant volume explosion pressure after a short induction time delay. The principal conclusions from the experiments of Apparatus 2 are that fully established detonations are formed very close to the wall where the maximum free radical concentration occurs within a distance of about a centimeter and that a radical concentration gradient appears necessary for the development of detonation. For very high flash intensities, where the radical concentration gradient is small and the entire explosion box is more or less uniformly excited and distributed with free radicals, constant volume explosions result. Thus, photo-chemical initiation requires both a mini-

mum peak concentration of free radicals to initiate a sufficiently fast reaction, as well as a gradient to enable a build-up of the pressure wave to form a detonation. Without a gradient, the entire excited volume simply explodes homogeneously after an induction delay.

With the interesting region in the immediate vicinity of the wall obstructed and not available for photographic observations in Apparatus 2, further schlieren observations were carried out in Apparatus 3. In Fig. 7, a series of schlieren photographs illustrate the formation of a deflagration wave when the flash energy is less than some critical value for direct initiation. It is observed that initially, a diffused roughly hemispherical region near the wall of incident·UV radiation begins to show signs of chemical reactions after the induction delay. A shock wave is then formed, as this hemispherical reacting volume grows. However, the shock wave is not sufficiently strong to cause auto-ignition and hence detonation. Thus, it decouples from the reaction front and a hemispherical deflagration is obtained. The phenomenon is identical to that of blast initiation in the sub-critical energy regime[8]. In the present case, the shock front is formed as a result of the chemical reaction of the gas mixture. In the blast initiation case, the shock is generated from the external energy release of the igniter. Fig. 8 shows the phenomenon corresponding to flash intensities exceeding the critical value. After an induction delay, the first frame illustrates that two fully established hemispherical detonation waves are already formed at the window. These two detonation waves are formed from two local "hot spots" at the window which may be due to some inhomogeneities. The time and spatial resolution of the present schlieren photography is insufficient to permit capturing the very rapid formation of these two hemispherical detonation waves. Considering that no external powerful

concentrated energy source is used, it is remarkable that a fully deve-
loped detonation can be formed within a distance of less than a centi-
meter relying solely on the basis of the chemical energy release of the
mixture itself. Since constant volume explosions can only result in a
shock wave of the order of $M_s \approx 2$, the radical concentration gradient
(or equivalently an induction delay gradient) must be responsible for
this extremely rapid amplification of the shock wave to form a detona-
tion. The subsequent frames of Fig. 8 show the coalescence of the two
hemispherical detonations to form a single multiheaded detonation wave
propagating at about the C-J velocity of the mixture.

It is to be noted that in all the present series of experiments
on photochemical initiation, the resultant detonation pressure is slightly
less than the C-J value. This is due to the fact that although detonation
is initiated at the window itself, the gas in the entire chamber has
been excited with some free radicals present due to photo-dissociation.
Thus as the detonation propagates into this slightly pre-dissociated mix-
ture, the strength of the detonation is slightly less than that accord-
ing to C-J calculation based on an initially undisturbed state. In Fig.
9, comparisons between the detonation pressures from photochemical ini-
tiation and from spark ignition in a 1 meter long tube are shown. It
can be seen that pre-dissociation lowers that C-J strength since with
pre-dissociation the shock can be weaker to give the same reaction rates
necessary to propagate a detonation.

4. Conclusion

The present study conclusively demonstrates that fully developed
detonations can be initiated directly (without going through the deflag-
ration regime) using flash photolysis. The phenomena of sub-critical and

critical flash intensities correspond to those of blast initiation where the igniter energy is sub-critical and critical respectively. However, for the very high super-critical flash intensities, the entire excited volume reacts approaching a constant volume explosion in constrast to blast initiation, where the very strong blast generated by the igniter decays progressively to a C-J wave in the super-critical energy regime. The present study indicates that the mechanisms of photo-chemical initiation are as follows: due to the exponential decay of free radical concentration away from the window of incident UV radiation, the layer of gas immediately next to the window has the shortest induction delay and explodes first. The shock wave generated then propagates into the next layer of gas which is first on the verge of exploding, since its induction delay is slightly longer than the layer next to the window. The shock then triggers the explosion and the resultant energy release enhances the shock and it becomes stronger as it advances to the next layer. This layer again is on the verge of exploding and the shock now triggers it and the subsequent energy release further enhances the shock. In this manner, the concentration gradient provides the means whereby energy can be released in phase with the shock. This coherent energy release gives rise to a very rapid amplification of the shock leading to the formation of detonation. This amplification of the shock wave by coherent energy release is analogous to the principle of a laser where stimulated emission triggered by the light wave provides coherent energy release to the light wave and gives rise to its amplification. Thus we may refer to to the mechanism of photochemical initiation as the SWACER mechanism representing "Shock Wave Amplification by Coherent Energy Release" in analogy to LASER which denotes "Light Amplification by Stimulated Emission of Radiation". The SWACER mechanism provides a means of generating very strong shock waves locally in a reacting medium without the use

of an external concentrated energy source. Thus the SWACER mechanism is
not restricted to photochemical initiation, and in fact there exists
strong evidence that it plays the principal role in the transition of
deflagration to detonation as well. Thus, in the pre-detonation deflag-
ration regime, the physical processes can be thought of as the agents
responsible for preparing the necessary conditions for the SWACER mecha-
nism to be operative.

References

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Figure Captions

WATER COOLING

QUARTZ TUBE

XENON FLASH TUBE

SILVER-COATED LASER CAVITY

NITROGEN FLOW

XENON FLASH TUBE

REFLECTOR

QUARTZ
WINDOW

SCHLIEREN
PHOTOGRAPHY

CYLINDRICAL EXPLOSION CHAMBER

QUARTZ WINDOW

XENON FLASH TUBE

Fig 3 Lee, Kapelovitz, Yoshikawa
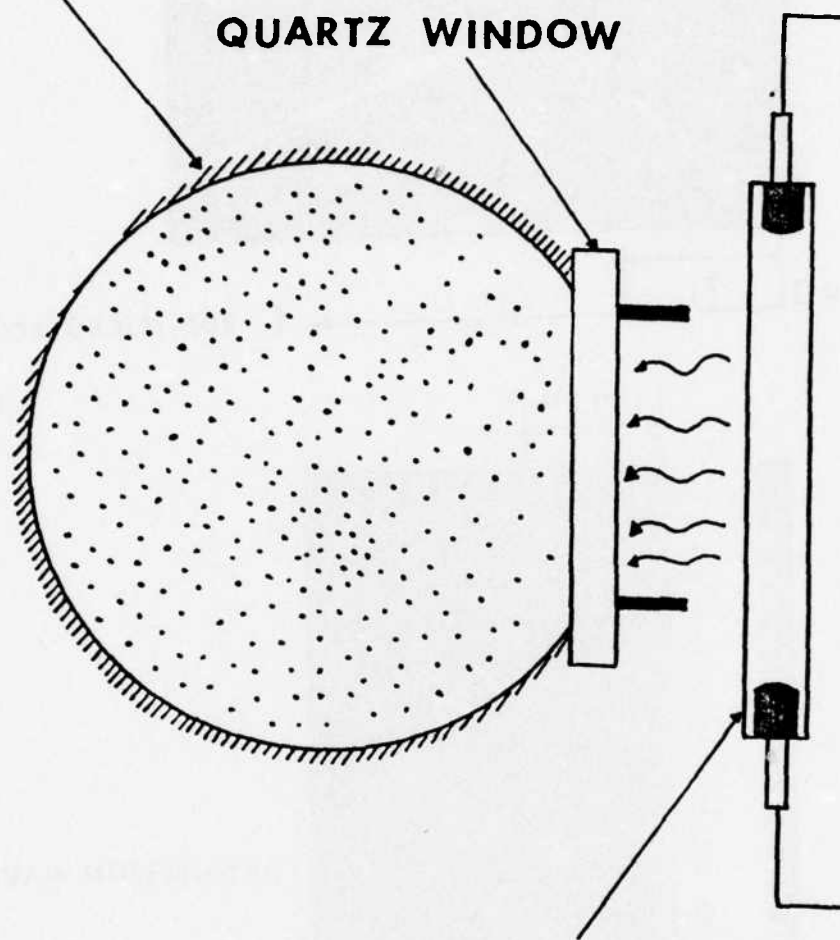
C₂H₂+O₂ 95% —NO₂ 5%   100 torr
FLASH ENERGY   6KJ

t=0   tᵢ   t 200 MICROSEC/DIV

50 CM

DETONATION WAVE

t=tᵢ

INDUCTION TIME

t=0
(START OF FLASH)

FLASH CAVITY

EXTENSION OF EXPLOSION TUBE

X=0
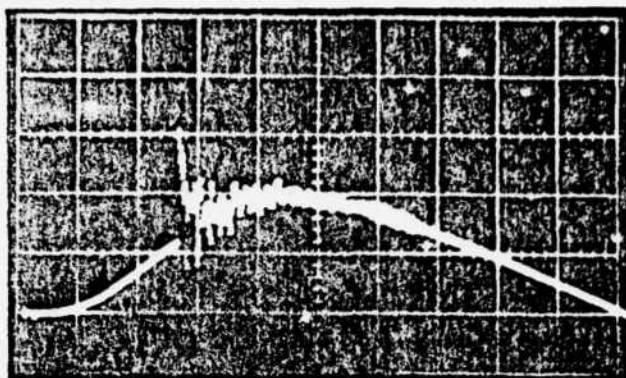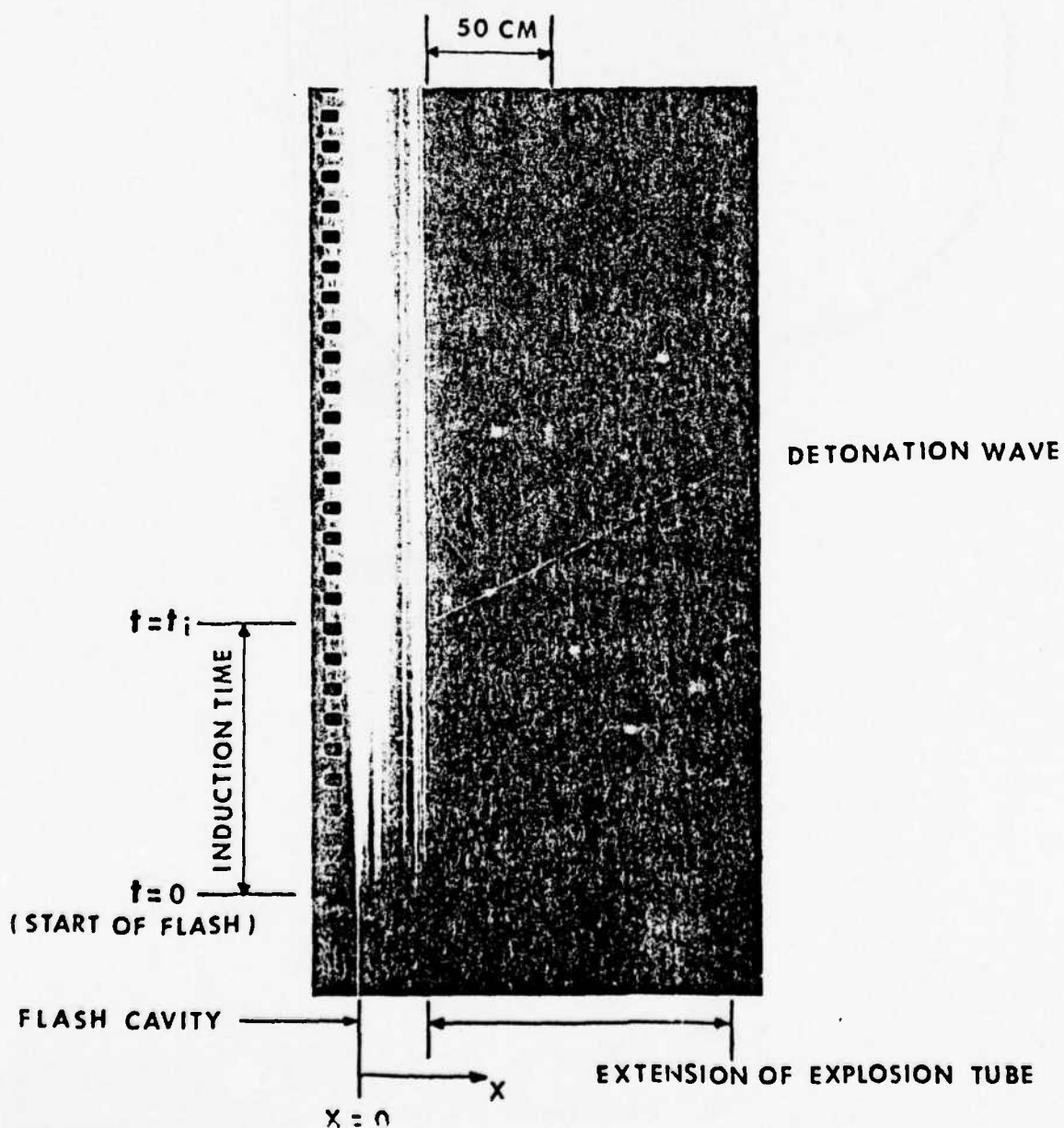
○ C_2H_2 50% - O_2 50%

⊖ C_2H_2 47.5% - O_2 47.5% - NO_2 5%

● C_2H_2 40% - O_2 40% - NO_2 20%

⊕ C_2H_2 30% - O_2 30% - NO_2 40%

△ H_2 50% - Cl_2 50%

(2.38 MSEC) 1 KJ ⟶○ ↑
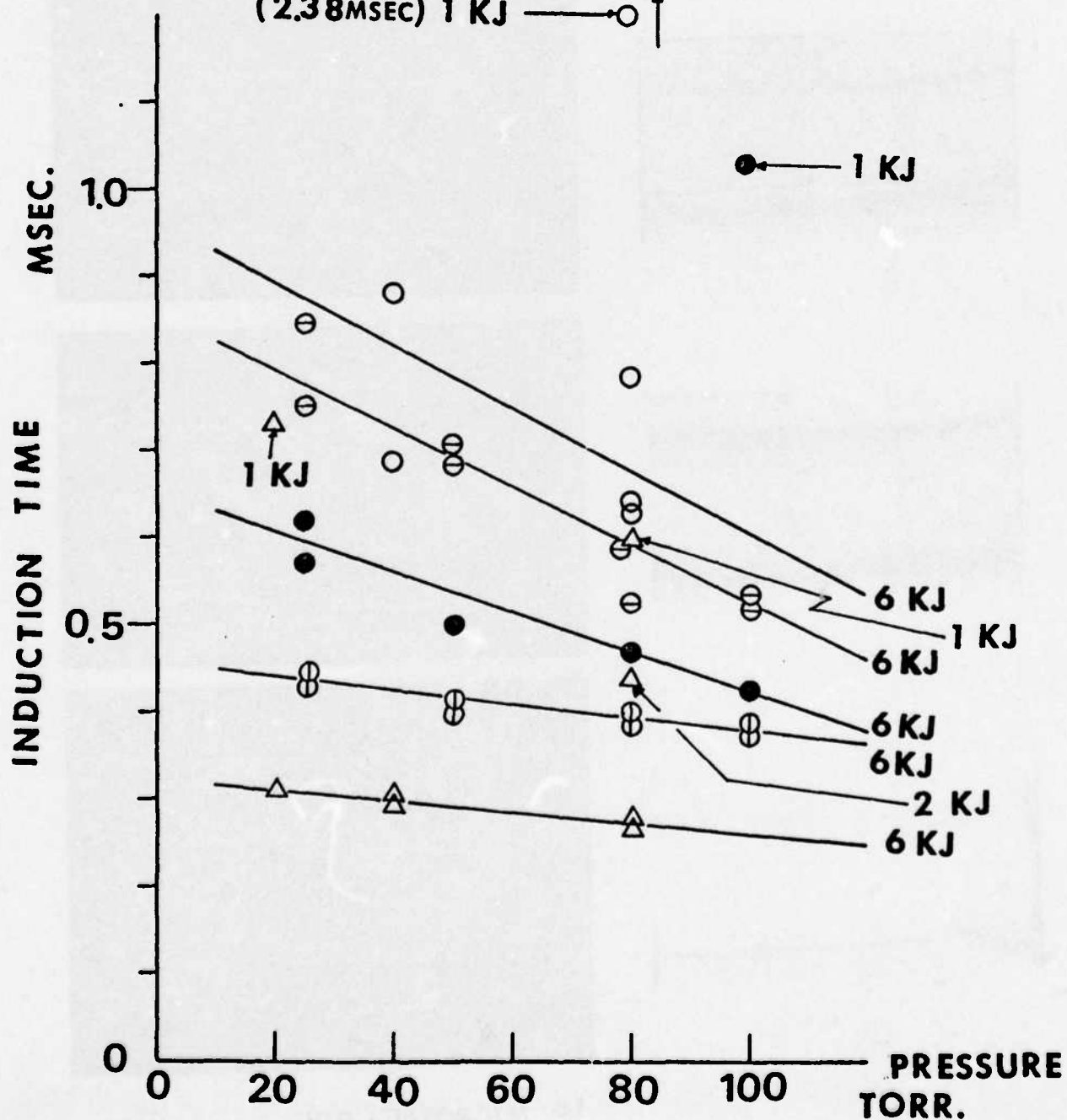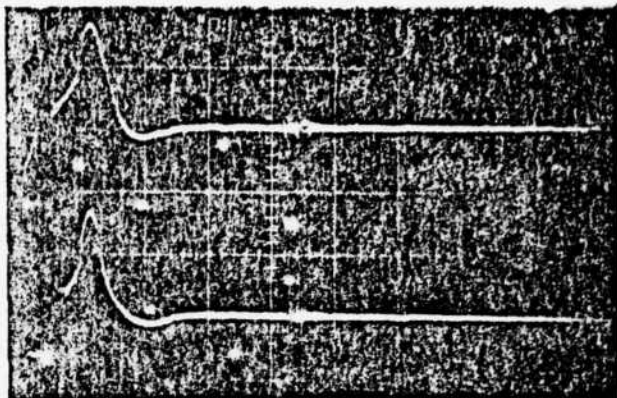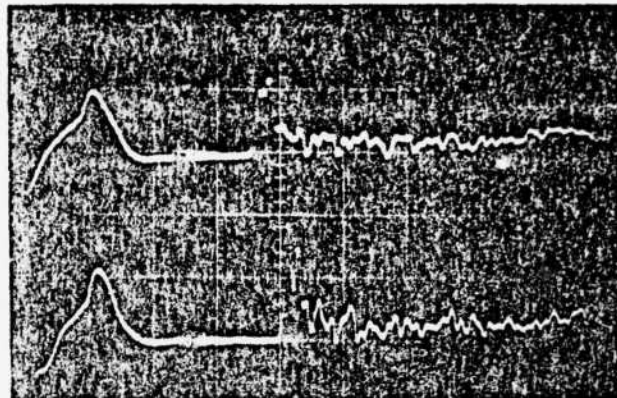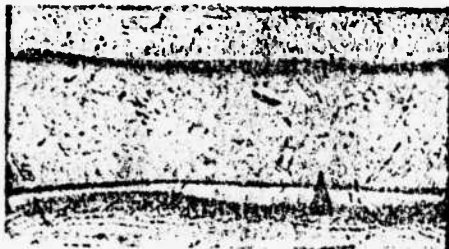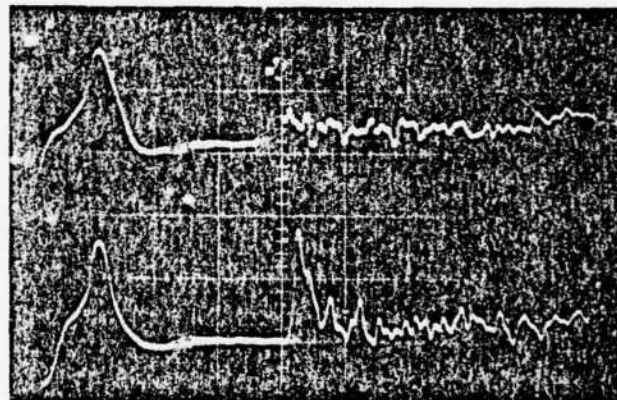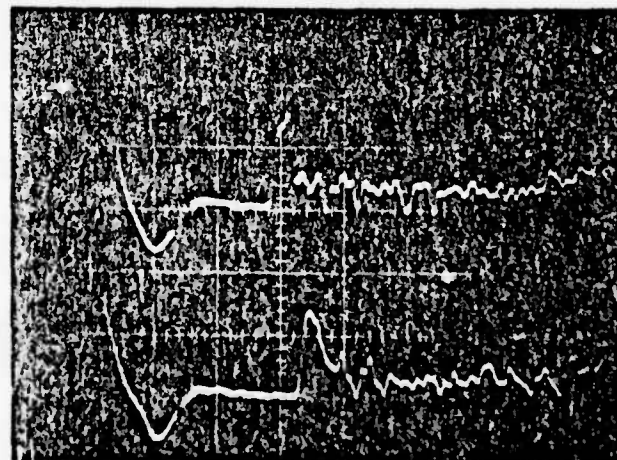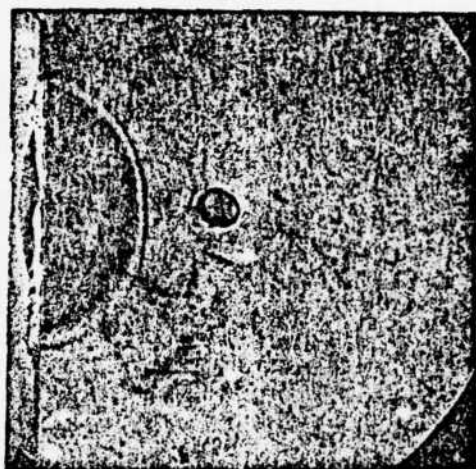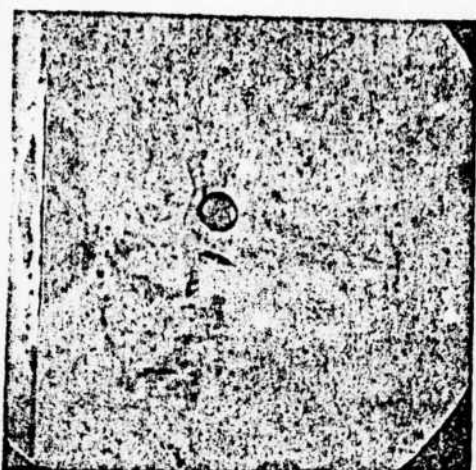
10 MICROSEC / DIV

5 CM
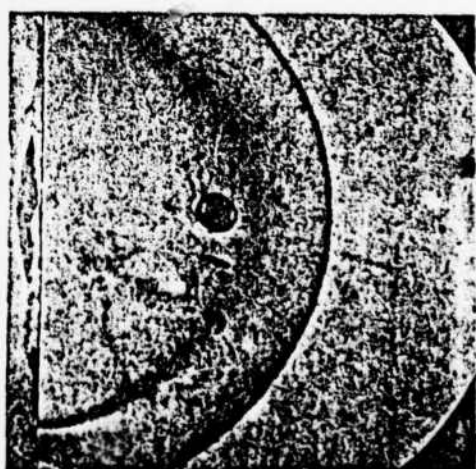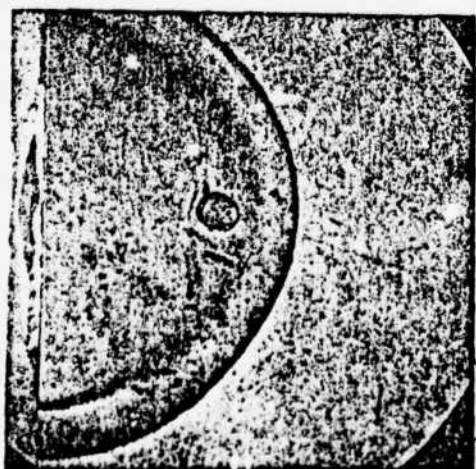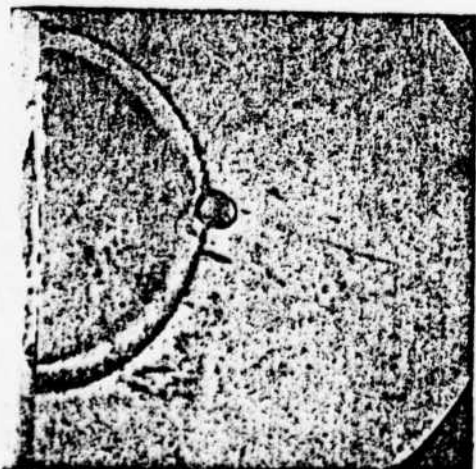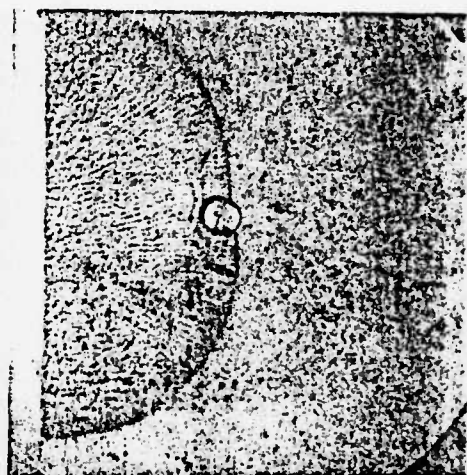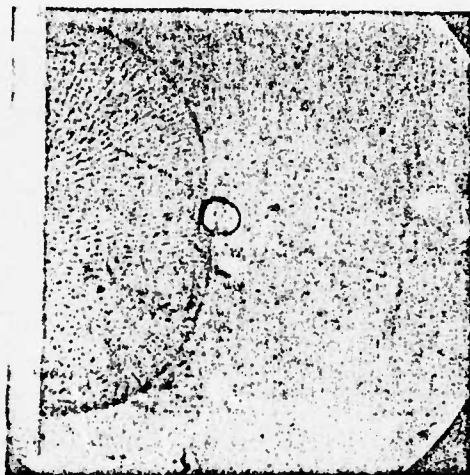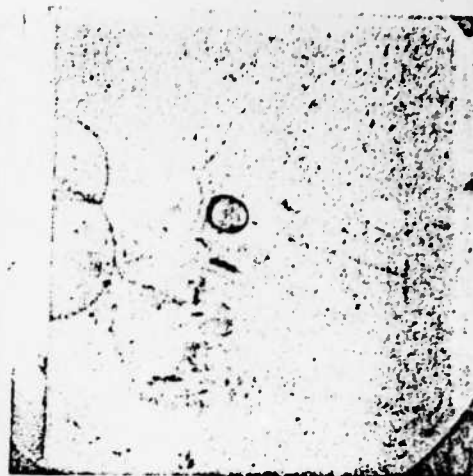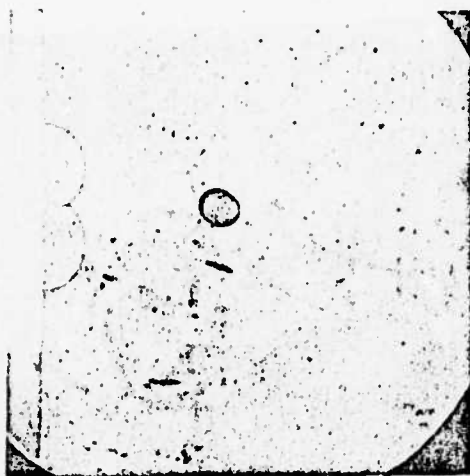
5 CM
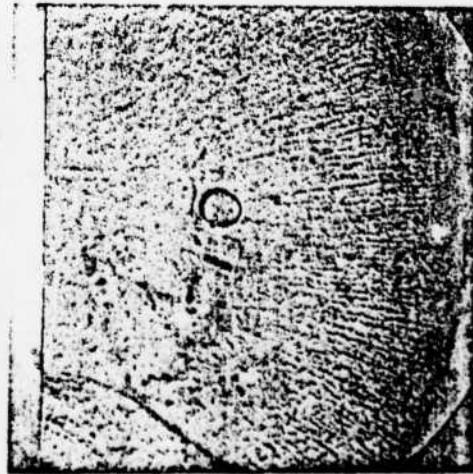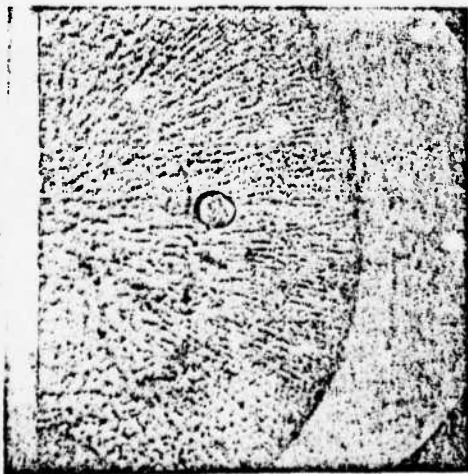
FLASH 8 MICROFARAD 12KV
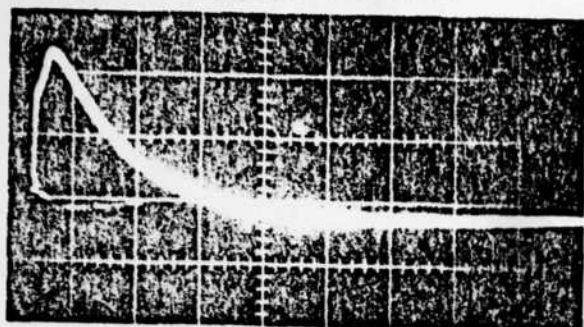
100 MICROSEC/DIV
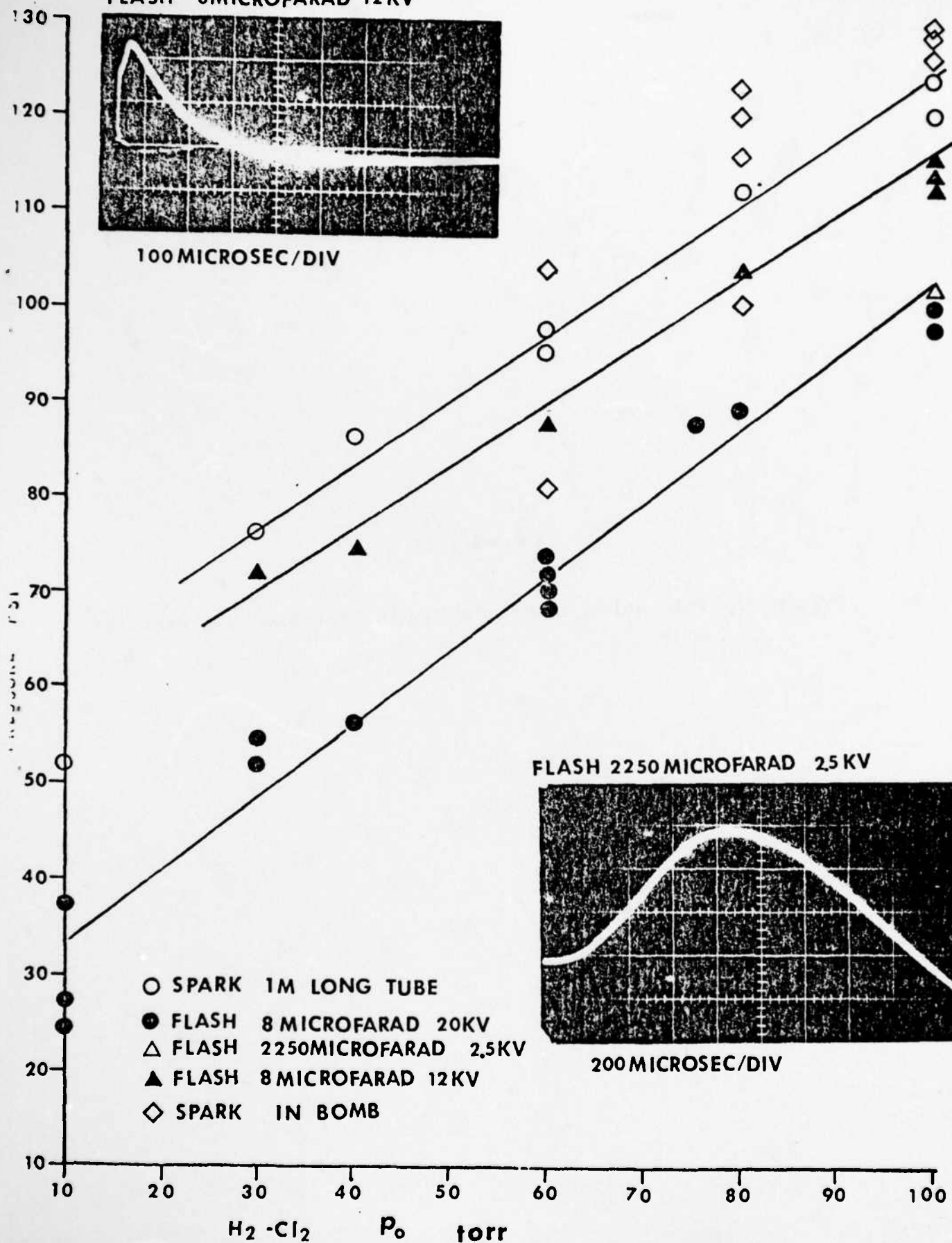
FLASH 2250 MICROFARAD 2.5 KV

200 MICROSEC/DIV

○ SPARK 1M LONG TUBE
● FLASH 8 MICROFARAD 20KV
△ FLASH 2250 MICROFARAD 2.5KV
▲ FLASH 8 MICROFARAD 12KV
◇ SPARK IN BOMB

H_2 -Cl_2 P_0 torr
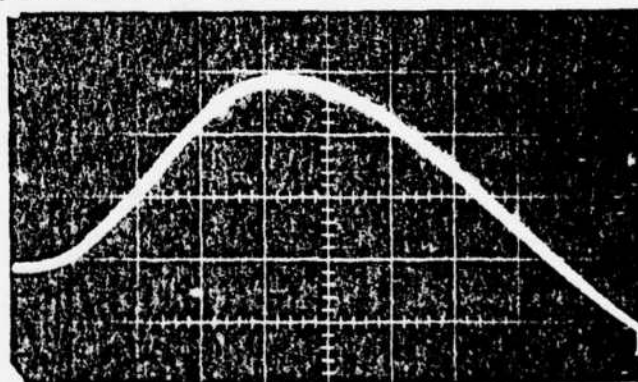
Appendix II

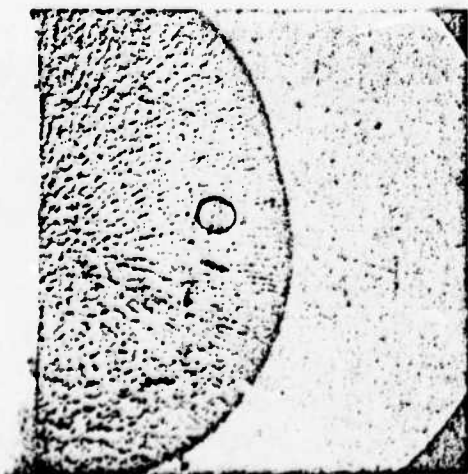Pyrophoric Initiation of Detonation in Explosive Gas Mixtures

Pyrophoric Initiation of Detonation in Explosive Gas Mixtures

by

R. Knystautas and J.H. Lee

A series of preliminary experiments were carried out to
investigate the feasibility of initiating a detonation wave in an
explosive gas mixture by catalytic means using the dispersion of
a pyrophoric material. This type of initiation scheme is consi-
dered to be the one most directly relevant to the development of
the single event third generation FAE device. It is envisaged that
in the operational version of such a device the initiating catalyst
would be dispersed with the fuel to mix with air to form an explo-
sive vapor cloud which would then detonate after an appropriate in-
duction time without further tactical intervention. In the present
feasibility study where we were looking at the mechanisms of this
mode of initiation, the pyrophoric material was force-injected in
gaseous form into an explosive gas mixture at sub-atmospheric ini-
tial pressure. The apparatus is illustrated schematically in Fig.
1 and consists of a piston driven compression cell connected to an
explosion tube which was about 20 cm in diameter and about 1 m long.
The compression cell was typically 1 cm in diameter and about 5 cm
long and was initially isolated from the explosion tube by a thin
polyethylene diaphragm. The piston was first pulled back all the
way from the diaphragm and then both the compression cell and explo-
sive tube were evacuated. The explosion tube was then filled with
equimolar oxy-acetylene gas mixtures to sub-atmospheric pressures
ranging from 100 to 200 torr. The compression cell was then charged
with gaseous pyrophoric material at slightly super-atmospheric

pressure. The pyrophoric material used was dimethyl zinc, $Zn(CH_3)_2$, whose boiling point is 46°C and whose vapor pressure is given as $\log_{10}P(torr) = 7.302 - 1560/T(°K)$. Thus to achieve a gaseous state of the pyrophoric material, the reservoir as well as the entire compression cell were wrapped with thermal tape and the entire assembly maintained at a temperature ∿65°C. The piston plunger was then activated by high pressure nitrogen and in accelerating into the DMZ charge, compressed it, ruptured the diaphragm and ejected a gaseous DMZ jet into the explosive gas charge. The combustion phenomena in the explosion tube were observed via self-luminous streak photography.

Typical streak records of pyrophoric initiation of detonation in equimolar oxy-acetylene mixtures at 125 and 200 torr and using injected DMZ are shown in Fig. 2. The initial luminous trajectory is that of the vortex ring traversing the length of the explosion tube. The chemical reactions in the intense mixing region of the vortex ring are not sufficiently strong to cause combustion of the explosive gas mixture. Thus about the first 500-1000 μsec or so correspond to the dispersion time of the pyrophoric material in the explosive gas mixture. Thereafter there is an incubation time of a further 500 μsec or so followed by the appearance of a detonation wave. It is to be noted that the detonation originates at the extreme end of the tube opposite to the injection orifice. This can be attributed to the fact that the pyrophoric jet traverses the length of the tube at these sub-atmospheric pressures and looses all its momentum at the end of the closed tube. Thus one would expect the highest catalyst concentrations at the end of the tube. It can be observed as expected that the induction time to

detonation decreases with increasing initial pressure, but that the dispersion time remains relatively invariant. Thus the feasibility of detonation initiation catalytically via the dispersion of pyrophoric material in an explosive gas mixture has been demonstrated. The mechanism of initiation here can be explained in terms of the SWACER mechanism where the dispersion in the transient ejected jet creates a gradient of concnetration of the catalyst. This in turn establishes the properly tailored gradient of induction time for the rapid amplification of shock waves via the SWACER mechanism. Current efforts are under way to achieve more detailed quantitative observations of this initiation technique. This will be achieved by ultra-high speed framing schlieren photography. Theoretical modelling of the SWACER mechanism is also being performed to achieve the necessary criteria for scaling up the present laboratory scale experiments to field tests and to the use of fuel-air mixtures rather than the more detonable fuel-oxygen mixtures of the present work.
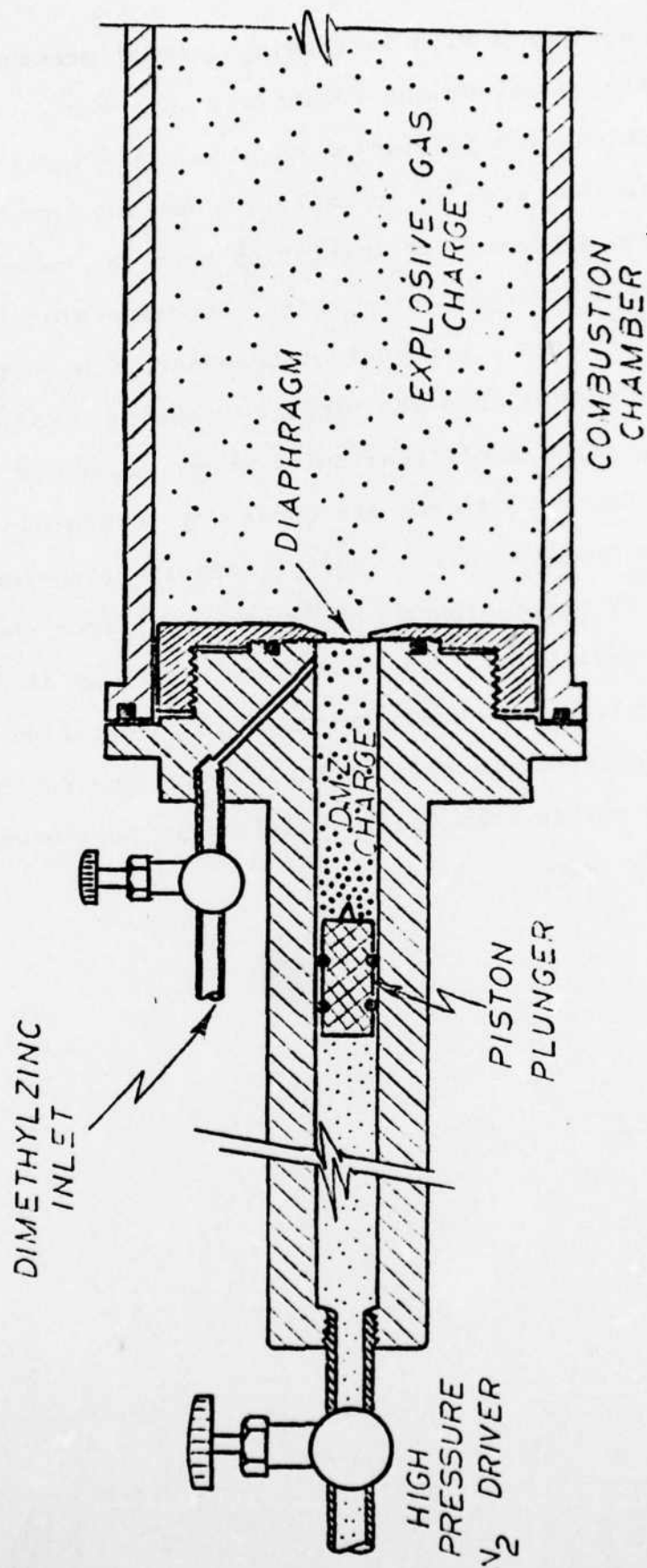
FIG. 1 PYROPHORIC INITIATION APPARATUS
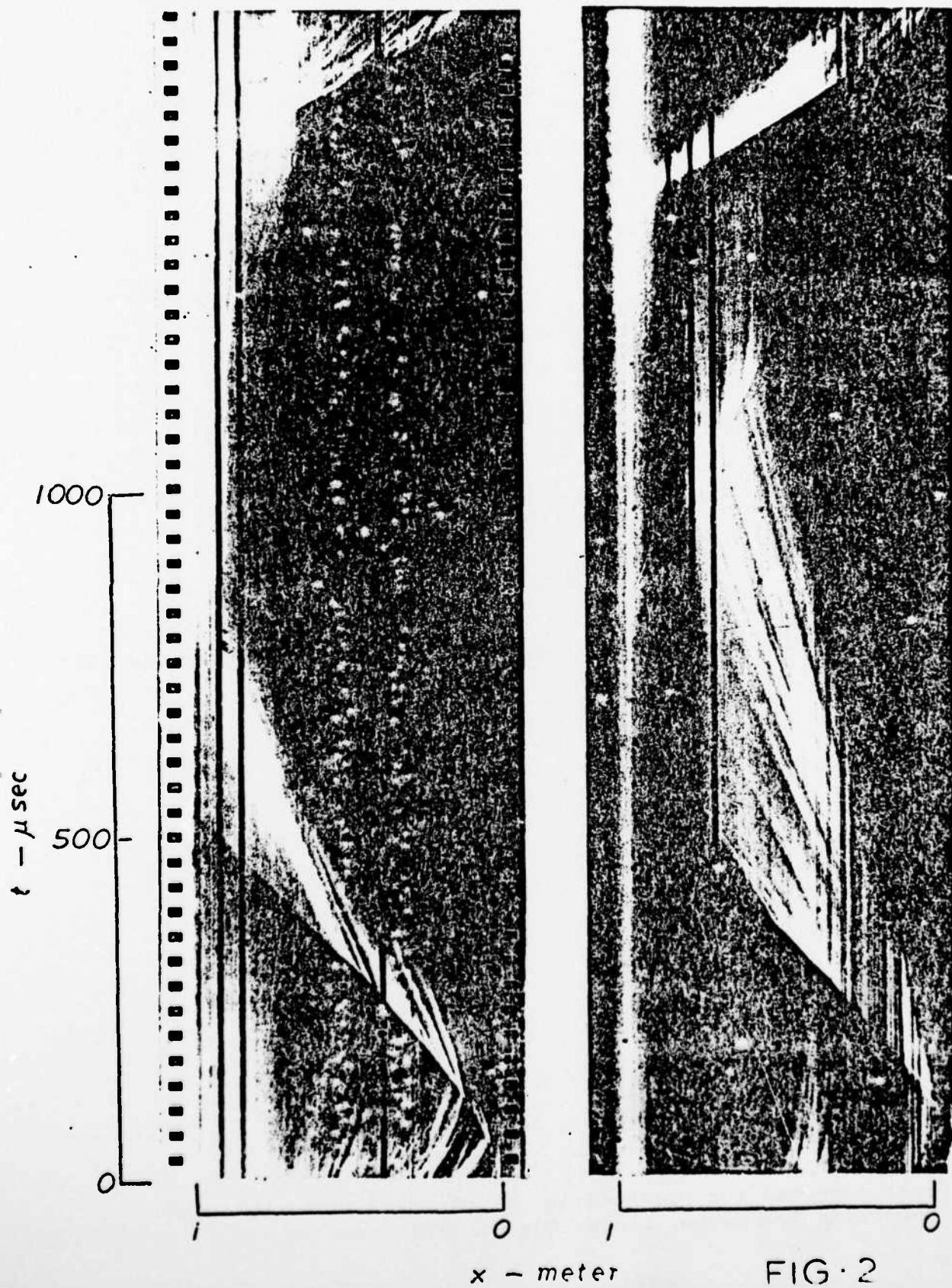
EQUIMOLAR $C_2H_2 - O_2$

125 torr   200 torr

$t - \mu\,sec$

1000

500

0

1   0   1   0

$x - meter$ FIG·2

Appendix III

Direct Initiation of Spherical Detonation by a Hot Turbulent Gas Jet*

Direct Initiation of Spherical Detonation by a Hot Turbulent Gas Jet

by

R. Knystautas, J.H. Lee, C. Guirao, M. Frenklach
McGill University, Canada

and

H.Gg. Wagner
University of Göttingen, W. Germany

Whether transition from deflagration to detonation can occur for a given fuel-air mixture is an extremely important question in connection with the safety in transport, storage and handling of large quantities of LNG and other flammables used in chemical industries. The urgency in obtaining an answer to this question need not be emphasized, but unfortunately the current state of the art in gaseous detonations does not permit this problem to be resolved with an acceptable degree of confidence. What is certain however are the requirements for detonation initiation, that is, a shock wave of sufficient strength to at least auto-ignite the mixture and with a duration of at least the induction period of this auto-ignition temperature must be generated. For most fuel-air mixtures where the auto-ignition temperature is of the order of 1500°K and an induction period at this temperature of about a millisecond, this means that a shock of $M_s \simeq 5$ and with a duration of about a millisecond must be generated if initiation of a detonation is to be achieved. For direct or blast initiation using a concentrated explosive charge, a strong shock of sufficient strength is always generated. Thus successful initiation depends on the duration which in turn depends on the charge weight. For transition from deflagration to detonation (also commonly referred to as DDT), the shock of the required strength and duration must be

generated via the energy release from the deflagration wave itself. It can readily be shown from simple calculations that phenomenally high flame speeds are required to generate the required strength shock of $M_s \sim 5$ and it is doubtful that the turbulence mechanism alone can give rise to such high flame speeds. Consider an alternate mechanism, that of packets of gases exploding instantaneously under constant volume conditions in a highly "folded" flame turbulent sheet. Simple shock tube calculations indicate that again only relatively weak shocks of $M_s \simeq 2$ are formed via this mechanism. Hence DDT especially under unconfined conditions is a highly unlikely process. On the other hand with partial confinement or with suitable obstacles in the path of the flame, the likelihood of DDT is greatly increased. In a recent experiment of Wagner[1], it was shown that upon passage of a hemispherical flame in slightly enriched ethylene-air mixture, a detonation is formed instantaneously downstream of the grid. It is not at all obvious how such strong shock waves can be generated by the flame upon transmission through the grid. In the present paper we report some recent results on an investigation of the mechanism of detonation initiation by a hot gas jet.

The experimental apparatus (Fig. 1) consists of a short flame tube 5 cm diameter by 20 cm long closed at the ignition end and with the other end terminated by an orifice plate. This tube is connected to a 15 cm diameter by 15 cm long cylindrical chamber at the center along an axis perpendicular to the axis of the cylinder. The ends of the cylinder are closed by two optical quartz flats to permit schlieren photography. The pressure transducers, one at the flame tube, the other at the cylindrical chamber register the combustion pressures developed. High speed spark schlieren

movies are used to observe the initiation phenomena as the hot
turbulent jet of product gases from the flame tube discharges
into the mixture in the cylindrical chamber. Round and two-dimen-
sional slit orifices of various sizes are used. In some experi-
ments, a wire grid is also placed across the orifice opening to
induce further turbulence in the jet. Acetylene-oxygen mixtures
diluted with various concentrations of N_2 are used and ignition
is via a weak electrical spark. A brief summary of the experi-
mental observations are as follows: The pressure record in the
flame tube (Figs. 2 and 3) demonstrates the usual smooth rise
and decay similar to that of a constant volume combustion process
depending on whether flame or detonation occured. The pressure
records in the larger cylindrical chamber show i) the smooth rise
and decay with sharp pressure spikes superimposed on it character-
istic of the shock flame interaction process in a closed chamber
or ii) the single sharp pressure peak of magnitude equal to the
reflected C-J detonation pressure. For a given mixture, initial
pressure, and type of orifice (i.e., circular, slit or circular
with grid) there exists a critical orifice opening where direct
initiation of spherical detonation is achieved by the hot turbulent
jet. Below or above a narrow range of orifice area about this
critical value, only a flame is observed in the big chamber. The
effectiveness in descending order of a particular type of orifice
in initiating a detonation is first the two-dimensional slit, then
the round orifice with wire grid, and finally just a round orifice.
The schlieren photographs reveal that upon ignition a jet of un-
burned mixture from the flame tube begins to discharge into the
larger cylindrical chamber due to the displacement by the flame.
The jet increases in velocity and reaches a maximum when the flame

reaches the orifice. A jet of hot combustion products now ejects downstream of the orifice. If the jet velocity and the turbulence intensity is low (large orifice opening) the mixing between the unburned gases and the hot combustion products which occurs at the interface is minimal. However, the contact with the hot product gases will ignite the unburned mixture and a flame develops in the jet. No detonation is initiated in this case and the pressure record in the cylindrical chamber registers a smooth pressure rise superimposed with spikes characteristics of pressure wave-flame interactions in a closed chamber. If on the other hand, the jet velocity and turbulent intensity is very high (from small orifice opening) then mixing at the interface is extremely rapid. The temperature drops to below the minimum required for ignition and no flame is ignited in the larger chamber. For a critical orifice opening, the mixing at the interface produces just the right conditions for the onset of detonation. After an ignition delay, typically of the order of 100 microseconds, an explosion occurs at some local hot spot in this turbulent mixing zone. The shock from this local explosion propagating in the hot gases of the mixing zone (which are just at the verge of auto-explosion themselves) accelerates rapidly and becomes a fully developed multi-headed detonation. A spherical detonation subsequently radiates outwards and pressure records as well as velocity measurements confirm that a C-J detonation is initiated. Typical schlieren photographs with accompanying pressure records are shown in Figs. 2 and 3. The importance of fine scale turbulence in the structure of the jet is indicated by the effectiveness of placing a wire grid over the circular orifice hole. Theoretical analysis is also made of the jet development from the flame tube with an orifice opening and the

jet velocities agree approximately with the experimental values of the order of 1000 m/sec. Using existing turbulent jet laws, the rate of entrainment of fresh mixture into the jet and the rate of temperature decrease of the jet can be calculated. An initiation criterion is proposed based on whether the induction delay for auto-ignition is within a prescribed value and agrees qualitatively with the experimental results. The important contribution of this paper is the demonstration that the conditions for the onset of detonation can be achieved via turbulent mixing at the interface between hot combustion products and unburned gases in a turbulent jet of sufficient fine scale turbulent intensity. This implies that partial confinement or obstructions in the path of a flame which can produce turbulent jets can lead to DDT in unconfined vapor clouds. The model developed in this paper also enables estimates to be made as to the possibility of DDT in a given mixture. Thus partial resolution of the important question can be achieved.

Reference

1. Dörge, K.J., Pangritz, D. and Wagner, H.Gg.; "Experiments on
    Velocity Augmentations of Spherical Flames by Grids", Acta
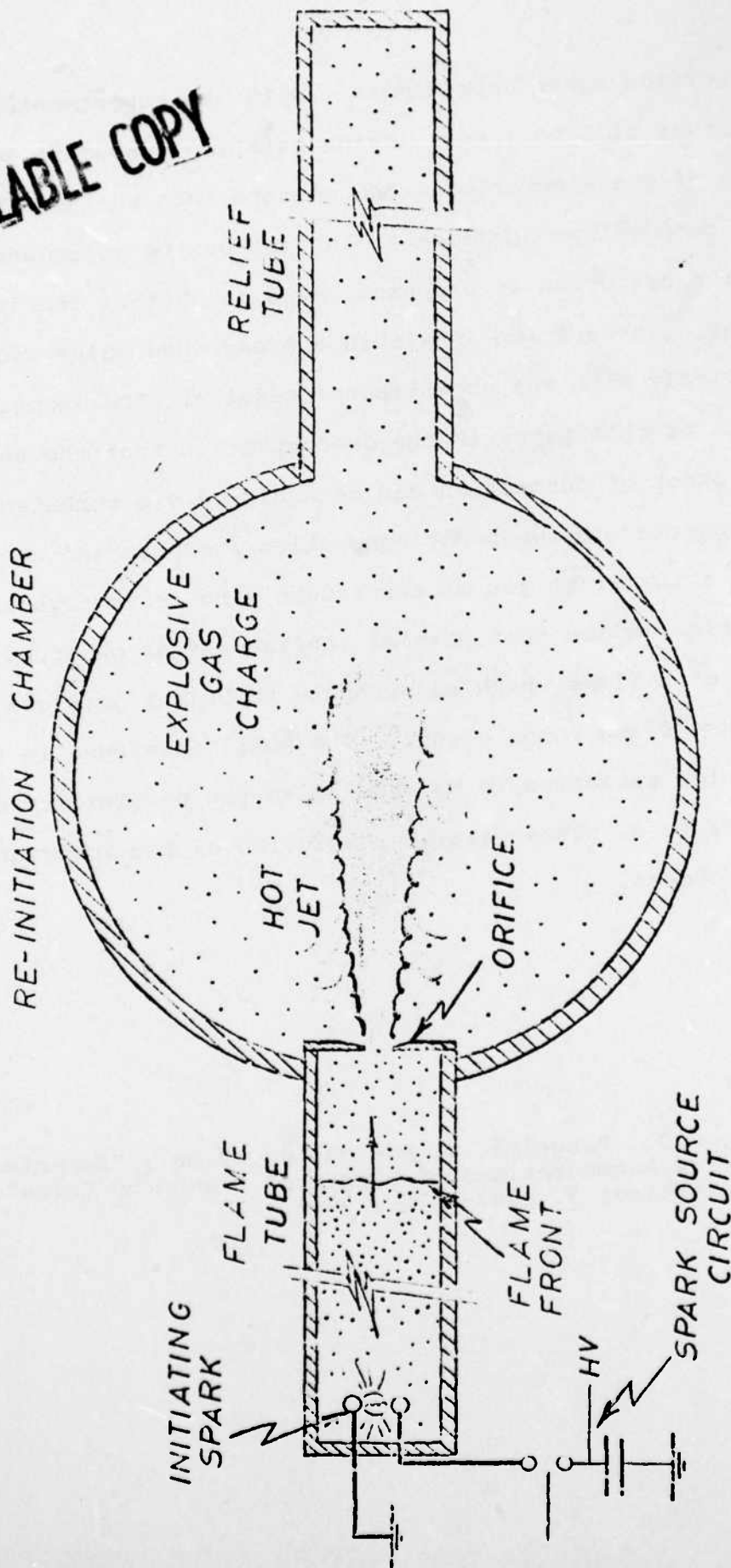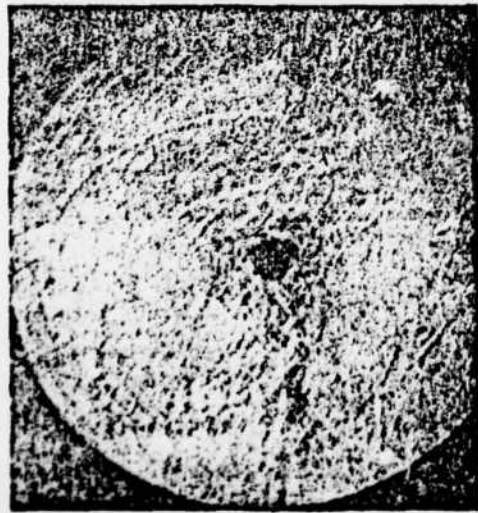   Astronautica, 3, 1067-1076 (1976).

FIG.1 HOT JET INITIATION OF DETONATION

NO DETONATION INITIATION
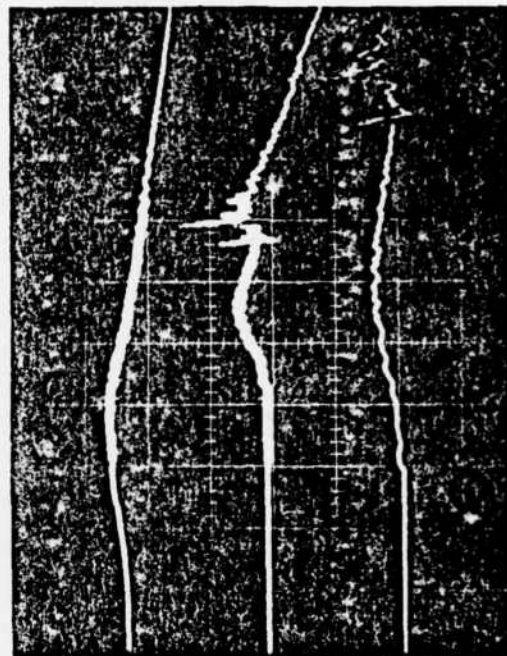
IGNITION

JET

PRESSURE UPSTREAM OF ORIFICE

PRESSURE DOWNSTREAM OF ORIFICE

500 μSEC/DIV

500 μSEC/DIV
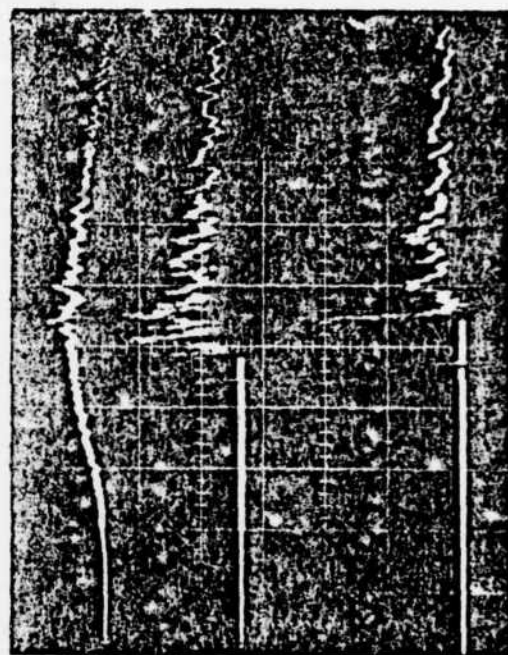
200 μSEC/DIV

FIG. 2
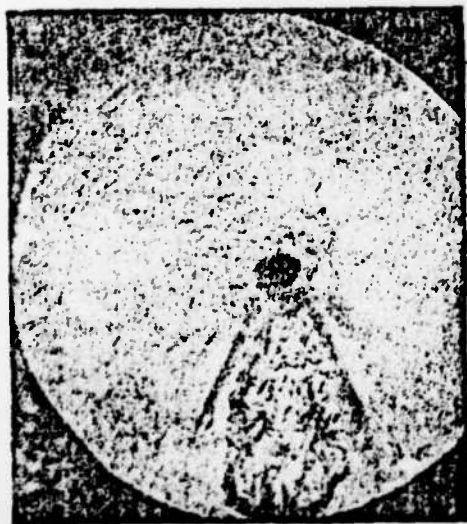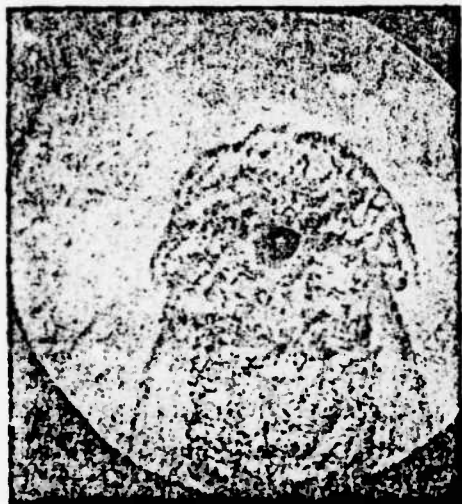
DETONATION GROWTH

DETONATION INITIATION

IGNITION

200 μSEC/DIV

200 μSEC/DIV

100 μSEC/DIV

PRESSURE UPSTREAM
OF ORIFICE

PRESSURE DOWNSTREAM
OF ORIFICE

FIG 3

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DIRECT INITIATION OF FUEL-AIR CLOUDS
DETONATION INITIATION
FAE III WEAPONS

20. ABSTRACT (Continue on reverse side if necessary and identify by block number)

During this period emphasis has been placed mostly on problems pertaining to feasibility of FAE III weapons. In particular major efforts have been devoted to the study of the mechanisms of initiation using various techniques other than the blast initiation method via concentrated explosive charges of conventional FAE weapons. Perhaps the most significant progress is the discovery of the universal initiation mechanism of Shock Wave Amplication by Coherent Energy Release (SWACER). When conditions for the SWACER mechanism to work have been generated locally in the explosive gas, then initially weak shocks can amplify extremely rapidly to form detonations in a time scale of

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223 165

the order of microseconds and corresponding length scales of the order of centimeters for the sub-atmospheric fuel-oxygen mixtures studied. It has been demonstrated experimentally that flash photolysis, intense turbulent mixing of either a pyrophoric compound (DMZ) or hot combustion products wth the explosive gas can lead to direct initiation via the SWACER mechanism. Theoretical modelling of the SWACER mechanism is being performed to achieve the necessary criteria for scaling up the present laboratory scale experiments to field tests and to the use of fuel-air mixtures rather than the more detonable fuel-oxygen mixtures of the present work.